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LONG-LIVED RADIOACTIVITY PRODUCED IN NORTHERN OHIO CONCRETE MATERIALS BY NEUTRON ACTIVATION

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SUMMARY

Measurements were made of trace element concentrations of concrete materials (cement, sand, and gravel) to be used in the NASA Plum Brook Space Propulsion Facility near Sandusky, Ohio. The purpose of the measurements was to select material that would minimize neutron activation of the completed facility during a nuclear test. Exposure rates resulting from best- and worst-case material selection are considered as a function of decay time after a nuclear test has been completed. The major activities found after neutron irradiation were due to cobalt 60, scandium 46, iron 59, lanthanum 140, and sodium 24. Cobalt 60 was significant only for irradiation times of an order equal to or greater than its half-life. For most projected nuclear tests, scandium 46 and iron 59 had the most significant activities. The ratio of cobalt to iron had a value of 0.00045 ± 0.00025 by weight for the materials tested.

INTRODUCTION

The Space Propulsion Facility is a large space simulation facility under construction at the NASA Plum Brook Station near Sandusky, Ohio (ref. 1). The test chamber, which is designed for high-vacuum operation, has a diameter of 100 feet (30.5 m) and a maximum height of 120 feet (36.6 m). The facility is designed for long-term endurance testing of large nuclear reactor space power systems at full power. The test chamber is surrounded by a 2-meter-thick biological shield of ordinary concrete with some borated areas to reduce neutron activation. Since personnel access inside the shield is required after reactor operation, it is desirable to keep the gamma ray exposure rate from neutron activation of materials inside the chamber low so that setups for later tests will not be hampered by radiation protection problems. It is also important that activation of the concrete shield (even its trace elements) not cause excessive exposure rate levels inside the chamber after nuclear testing. Concrete materials (cement, sand, and large aggregate) have trace element concentrations characteristic of the local area from which they are obtained; thus, it is necessary to measure materials from the local area rather than rely on literature values. Therefore, a construction materials program was started to measure concentrations of trace elements which produce long-lived radioactivity through neutron capture in concrete. Neutron irradiation was used instead of simple chemical analysis in order to avoid overlooking the type of trace element with a high cross section but low concentration that could still produce high activities in the finished concrete. Both preliminary measurements and data on the actual construction materials used are given.

CONCRETE MATERIALS SPECIFICATIONS

Preliminary measurements of neutron activation of locally available concrete materials were used to set specifications which would result in the use of the best locally available materials, subject to some economic constraints. The trace element specifications are given in table I. The column entitled "Other" lists those long-lived activities (a few days or longer in half-life) which might appear unexpectedly in the concrete materials being tested. These long-lived activities approximate the activity of cobalt 60 in the same material after a long-term exposure.

A distinction was made in the specifications for two types of sand. Sand dredged from Lake Erie, although considerably higher in trace elements than that made from crushed limestone, was easier to handle in the low-water- and low-cement-content concrete used. Therefore, Lake Erie sand was used in areas where activation was less of

TABLE I. - TRACE ELEMENT SPECIFICATIONS FOR CONCRETE MATERIALS

USED IN SPACE PROPULSION FACILITY

Material	Trace element			
	Cobalt, ppm by weight	Scandium, ppm by weight	Iron, percent by weight	Other, $\frac{\text{disintegrations}}{\text{(sec)(kg)}}$ (a)
Cement	30	20	6	5.7×10^{3}
Sand for borated and nonsurface area concrete	20	20	12	3.8
Sand for nonborated dome surface concrete	3	4	4	.57
Number 4 stone	3	4	4	.57
Number 12 stone	3	4	4	.57

[a]Maximum saturated activity for thermal neutron flux of 10^{6} neutrons/(cm^{2})(sec).

TABLE II. - BIOLOGICAL SHIELD CONCRETE MIX FOR SPACE PROPULSION FACILITY

Material	Concentration	
	lb/cu yd	kg/cu m
Water	150	89.0
Cement	370	220
Sand	1050	623
Large aggregate	2580	1530
Water-reducing agent	.2	.1
Air-entraining agent	.8	.5

a problem, such as in borated concrete. Table II gives the actual mix used. The density of the concrete was 142 to 152 pounds per cubic foot (2270 to 2440 kg/cu m). A low-cement- and low-water-content concrete was chosen to minimize shrinkage. In borated concrete, boron frit was used to replace part of the sand.

NEUTRON ACTIVATION MEASUREMENTS

Samples of cement, sand, and large aggregate were taken on the basis of one sample per 100 to 500 tons (90 000 to 450 000 kg) of material. The sampling rate varied with the material to be tested and the size of the stockpile. The samples were ground to powder, and 0.020 kilogram weighed out for reactor irradiation. After weighing, the samples were put in double polyethylene bags, and each bag was heat-sealed separately. The irradiations were performed in rabbit-tube facilities at the Plum Brook Reactor Facility, Sandusky, Ohio, or at a contractor's reactor research facility. The thermal neutron fluences (time-integrated fluxes) for the irradiations were 10^{14} to 10^{15} neutrons per square centimeter. Higher fluences would have made the analysis easier; however, some trouble was experienced with plastic bag breakage at the higher levels, presumably due to gas generation. As expected, it was necessary to allow short-lived activities to decay before beginning the analysis of long-lived ones. After 1 to 2 hours (3600 to 7200 sec), the only short-lived activity of any significance left was due to sodium. Because of the relatively short half-life of sodium 24 (table III) and its resulting activity predominance, it was necessary to wait several days before gamma spectroscopy of long-lived activities could be performed. Table IV gives trace element concentrations present in

TABLE III. - RADIATION CHARACTERISTICS OF VARIOUS TRACE ELEMENTS AFTER NEUTRON IRRADIATION

Element	Isotope produced	Half-life		Gamma ray energy, MeV
Cobalt	Cobalt 60	5.3 yr	1.7×10^{8} sec	1.17, 1.33
Scandium	Scandium 46	84 days	7.3×10^{6} sec	0.89, 1.12
Iron	Iron 59	45 days	3.9×10^{6} sec	1.10, 1.29
Sodium	Sodium 24	15 hr	5.4×10^{4} sec	1.37, 2.75
Lanthanum	Lanthanum 140	40 hr	1.4×10^{5} sec	0.33, 0.49, 0.81, 1.6
Zinc	Zinc 65	245 days	2.1×10^{7} sec	1.1

TABLE IV. - AVERAGE TRACE ELEMENT CONCENTRATIONS IN CEMENT, SAND, AND STONE USED IN SPACE PROPULSION FACILITY CONSTRUCTION

Material	Cobalt, ppm by weight	Scandium, ppm by weight	Iron, percent by weight	Sodium, ppm by weight	Lanthanum, ppm by weight
Cement, type II	8.0±0.6	4±1	3.2±0.2	----------	36±4
Lake Erie sand	14.3±0.6	6±2	5.9±0.2	10 300±7100	55±7
Crushed limestone sand[a]	3.9±0.2	1.1±0.3	.69±0.04	530±40	32±3
Number 4 limestone[a]	2.6±0.2	1.1±0.3	.73±0.02	340±240	26±2
Number 12 limestone[a]	2.8±0.3	1.2±0.4	.72±0.02	520±190	27±2
Bulk limestone[b]	1.5±0.5	2.6±0.7	.42±0.15	185±33	54±9

[a]Quarry 1.
[b]Quarry 2.

TABLE V. - TRACE ELEMENT CONCENTRATIONS IN MISCELLANEOUS CONCRETE MATERIALS

Material	Isotope for which activity was measured (a)	Element concentration, percent by weight
Boron frits	Zinc 65	1.2 Zinc
Pozzolan	Zinc 65	0.07 Zinc
White cement, type I	Iron 59	1.1 Iron
	Scandium 46	1.3 Scandium (ppm)
Gray cement, type I	Iron 59	6.1 Iron
	Scandium 46	2.4 Scandium (ppm)
High-early-strength cement, type III	Iron 59	5.8 Iron
	Scandium 46	2.2 Scandium (ppm)

[a]Cobalt was not measured because of its long half-life and consequent low count rate. It is probably present in cements along with iron (tables VI and VII).

cement, sand, and large aggregate. (All errors given are based on 90 percent confidence limits.) Table V gives the results of some preliminary measurements on other local concrete materials. With the exception of the boron frits, these materials were not used in Space Propulsion Facility construction. The pozzolanic material is the chemical calcium lignin sulfonate. It is used to improve concrete fluidity during pouring and is normally used at the rate of 1 to $1\frac{1}{2}$ pounds per cubic yard (0.6 to 0.9 kg/cu m) of concrete. Trace element concentrations in water were not measured; the activity from water is generally small (see, e.g., scandium content of water, table IX, p. 8). Standard chemical techniques were used, in addition to neutron activation, for iron measurements.

TABLE VI. - COBALT-IRON RATIOS IN CONCRETE MATERIALS

Material	Weight ratio of cobalt to iron
Cement, type II	0.00025
Lake Erie sand	.00024
Crushed limestone sand from Sandusky, Ohio, area[a]	.00057
Number 4 limestone from Sandusky, Ohio, area[a]	.00036
Number 12 limestone from Sandusky, Ohio, area[a]	.00039
Bulk limestone from Sandusky, Ohio, area[b]	.00036
Limestone A from Southern Calif.[c]	.00020
Limestone B from Southern Calif.[c]	.00057
Limestone C from Southern Calif.[c]	.00072
White Cement from Southern Calif.[c]	.00035
Igneous rocks[d]	.00040
Average	0.00045±0.00025

[a]Quarry 1.
[b]Quarry 2.
[c]Ref. 2.
[d]Ref. 4.

COBALT-IRON RATIO

There appears to be a correlation between the iron and cobalt contents of the various materials tested, as might be expected from their similar chemical characteristics. Table VI gives ratios of cobalt to iron in the materials tested, as well as in a wide range of other concrete materials. The average cobalt-iron ratio is given by $C_{Co}/C_{Fe} = 0.00045 \pm 0.00025$, where C is concentration by weight. The cobalt activity is normally much lower than the iron activity for short irradiations because of the long half-life of the cobalt 60. This, plus the fact that the cobalt and iron gamma ray peaks are close together (table III), makes it very difficult to separate the two activities by gamma ray spectrometry. The observed correlation of cobalt and iron contents permits estimation of cobalt content from, say, wet chemical analysis of iron content. Table VII

TABLE VII. - TRACE ELEMENTS OF CONCRETE MATERIALS IN SANDUSKY, OHIO, AREA RELATIVE TO IRON

Material	Cobalt	Scandium	Sodium	Lanthanum
	Concentration relative to iron			
Cement, type II	2.5×10^{-4}	1.3×10^{-4}	----	11×10^{-4}
Lake Erie sand	2.4	1.0	0.17	9
Crushed limestone sand[a]	5.7	1.6	.08	46
Number 4 limestone[a]	3.6	1.5	.05	36
Number 12 limestone[a]	3.9	1.7	.07	38
Bulk limestone[b]	3.6	6.2	.04	129

[a]Quarry 1.
[b]Quarry 2.

gives trace element concentrations relative to iron in various concrete materials. It should be noted from table VII that the cobalt-iron ratio varies to a lesser extent than any of the ratios of other trace elements to iron.

TOTAL CONCRETE ACTIVATION

It is difficult to calculate a resultant total concrete activity which is representative of concrete used for the Space Propulsion Facility because different combinations of materials are used at different locations. Therefore, some best and worst activation results are presented for the mix proportions given in table II (p. 3).

TABLE VIII. - ASSUMED TRACE ELEMENT CONCENTRATIONS FOR BEST AND WORST CONCRETE

Trace element	Best concrete	Worst concrete	Worst/best
	Concentration by weight		
Cobalt	2.6 ppm	6.0 ppm	2.3
Scandium	1.3 ppm	3.5 ppm	2.7
Iron	0.7 percent	2.2 percent	3.2
Sodium	250 ppm	3200 ppm	12.8
Lanthanum	27 ppm	51 ppm	1.9

For the best concrete, data on type II cement, crushed limestone sand, and the lowest activity large aggregate were considered. For the worst concrete, data on type II cement, Lake Erie sand, and the highest activity large aggregate were used. Table VIII lists the resulting trace element concentrations, and figure 1 shows the corresponding dose rate at a distance of 1 meter from a 1-kilogram sample after a 90-day (7.8×10^6 sec) exposure period in a unit thermal neutron flux. The 90-day (7.8×10^6 sec) irradiation time is considered representative of testing a large nuclear-electric space power system.

Design calculation results (ref. 3) for a 90-day run of an unshielded 600-thermal-kilowatt reactor operating near the chamber center give a gamma exposure rate of 150 milliroentgens per hour to personnel on the chamber floor; the calculations include a 1-week cooling period after reactor shutdown. Since experimental activation data corresponding to worst-case concrete in figure 1 are roughly comparable with values assumed in the design calculation, a normalized exposure rate of about 4×10^{-14} roentgen per hour in figure 1 corresponds approximately to an exposure rate of 150 milliroentgens per hour in the chamber from concrete activation alone. Furthermore, since surface boration of the concrete shield will reduce the exposure rate by a factor of about 5 (ref. 3), it is not anticipated that concrete-trace-element activation will produce personnel access problems if decay times of several days to a week are allowed.

Figure 1 shows that, while sodium 24 is the major long-lived isotope at reactor shutdown, scandium 46 and iron 59 will be the predominant activities for 1 week (6.1×10^5 sec) to 2 months (5.3×10^6 sec) after testing; the activity of the scandium 46 will continue to predominate for about 6 months (1.6×10^7 sec). After about 1 year (3.2×10^7 sec),

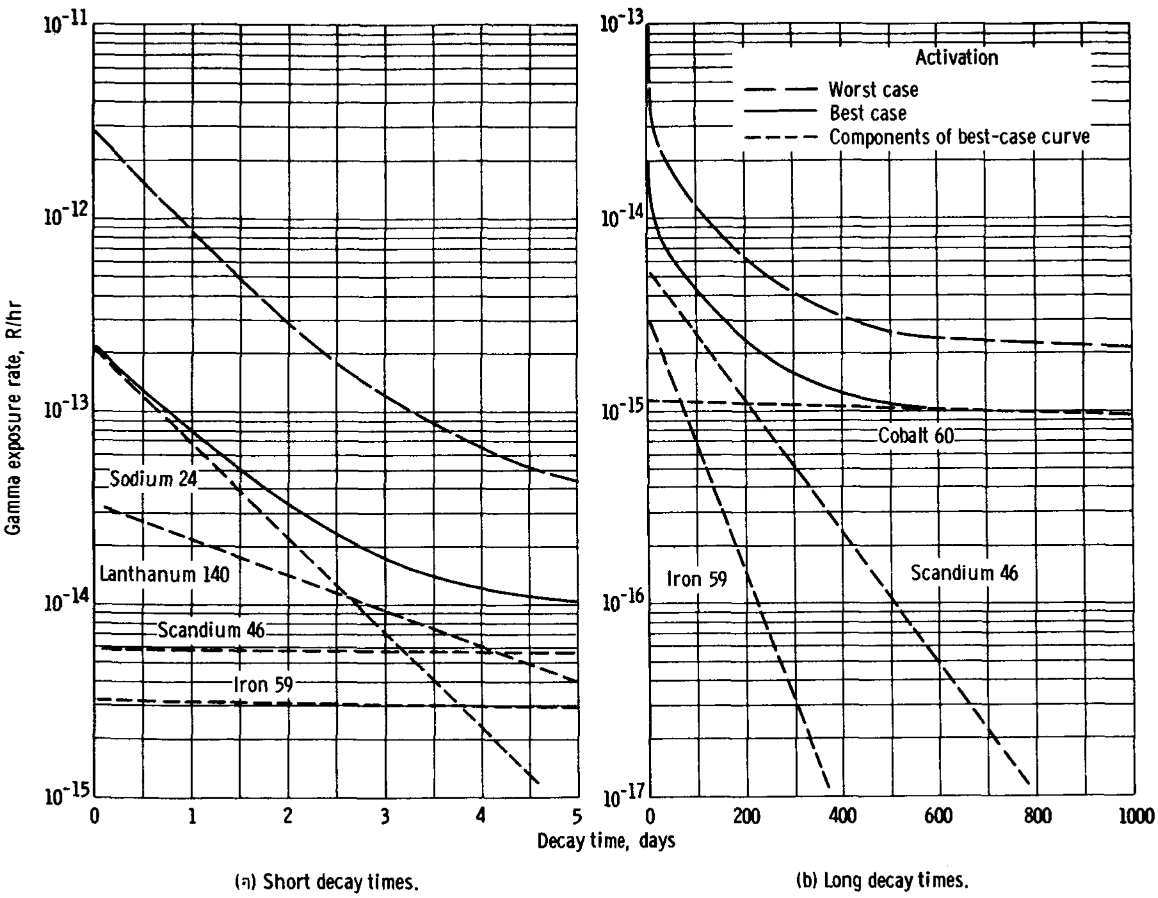

Figure 1. - Concrete gamma exposure rate for 90-day thermal neutron irradiation period. Thermal neutron flux, 1 neutron/cm^2-sec; sample weight, 1 kilogram; distance from sample, 1 meter.

cobalt 60 has the major residual activity. It is also of interest to note that, while the worst activation dose rate is more than a factor of 10 higher than the best rate at shutdown, this difference reduces to a factor of 3 after 1 week (6.1×10^{5} sec), since the widest variation in concentration from best to worst occurs for sodium (table VIII).

SCANDIUM

Scandium 46 was one of the major activities in the concrete materials tested. Even so, the scandium content of limestone from the Sandusky, Ohio, area (1.1 to 2.6 ppm) is significantly less than the average concentration of scandium in the Earth's crust (6 ppm).

TABLE IX. - SCANDIUM IN VARIOUS MATERIALS

Material	Scandium concentration, ppm by weight
Cement, type II	4±1
Lake Erie sand	6±2
Limestone from Sandusky, Ohio, area[a]	1.1±0.3
Limestone from Sandusky, Ohio, area[b]	2.6±0.7
White cement, type I	1.3
Gray cement, type I	2.4
High-early-strength cement, type III	2.2
Earth crust average[c]	6
Malay archipelago rocks[c]	2.7
Igneous rocks in general[c]	0.7 to 65
Metamorphic rocks[c]	0.7 to 100
Sedimentary rocks[c]	2 to 100
Wolframite[c]	Up to 1400
Sea water[c]	4×10^{-5}
Water with high bicarbonate ion[c]	.06
Meteorites (chondrites)[c]	4.5 to 10
Clays and shales[d]	10
Norwegian rocks and soils[e]	11 to 21
California limestone[f]	0.03 to 0.12
California white cement[f]	1.36
Beryl[c]	2000

[a]Quarry 1.
[b]Quarry 2.
[c]Ref. 5.
[d]Ref. 6.
[e]Ref. 7.
[f]Ref. 2.

However, scandium can exist in greater and lesser concentrations (table IX). Tests of California limestone (ref. 2) show it to have the lowest scandium content of any Earth-crust material for which data were available.

SUMMARY OF RESULTS

Trace element concentrations were measured in concrete materials from the Sandusky, Ohio, area. Neutron activation was combined with gamma ray spectrometry and chemical separation for most of the measurements. Iron content was measured by standard chemical methods as well as by neutron activation. The following results were obtained:

1. Proper selection among local concrete materials can reduce radiation exposure from concrete shield activation by a factor of 10 for decay times less than 1 day and by a factor of 3 for decay times greater than 1 week.
2. The principal long-lived activities found after neutron activation of cement, sand, and gravel were sodium 24, lanthanum 140, iron 59, scandium 46, and cobalt 60.
3. Scandium 46, although not usually mentioned in similar measurements on concrete from other locations, was a major contributor to the total activity.
4. The cobalt-iron weight ratio for a wide range of materials had an average value of 0.00045 ± 0.00025. Thus, an iron content measurement for the same class of materials allows an estimate of cobalt concentration within a factor of 2.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, July 6, 1966,
120-27-01-03-22.

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